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Supporting information

Poly(ionic liquid)s with Branched Side Chains: Polymer Design for Breaking Conventional Record of Ionic Conductivity

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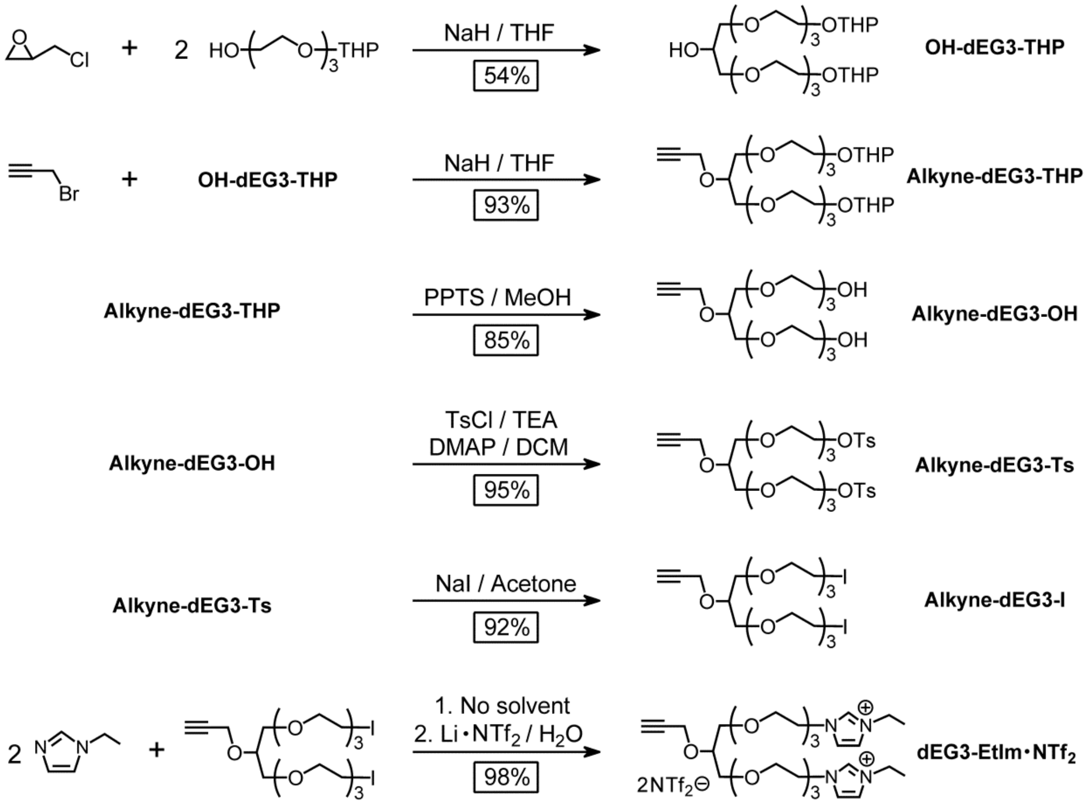
1.1. Synthesis of di-branched cationic alkyne derivative



Scheme S1. Synthesis of di-branched cationic alkyne derivatives

OH-dEG3-THP: THP-protected triethylene glycol[S1] (12.0 g, 51.2 mmol) was dissolved in dry THF (40 mL). NaH (2.2 g, 60 wt% in mineral oil) was added portionwise with stirring (*Caution: Careful for hydrogen and heat generation*). The reaction mixture was stirred at room temperature for 30 min under N_2 atmosphere. After adding epichlorohydrin (1.8 mL, 23 mmol), the reaction mixture was stirred at 60 ºC for 24 h. After cooling to room temperature, CH_2Cl_2 (200 mL) was added. The organic solution and 0.1 N $NaHCO_3$ aqueous solution (100 mL) were shacked with a separation funnel. The organic layer was collected and dried with $MgSO_4$, filtered and concentrated by evaporation. The crude product was purified by column chromatography (SiO_2; Hexane/Acetone = 1/1). Yield: 6.5 g (54 %). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.45−1.90 (m, 12H), 3.20 (d, J = 4.4 Hz, 1H), 3.45−3.73 (m, 28H), 3.80−3.90 (m, 4H), 3.96 (m, 1H), 4.63 (t, J = 3.6 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ =19.6, 25.6, 30.7, 62.4, 66.8, 69.5, 70.6, 70.7, 70.8, 70.9, 72.7, 99.1.

Alkyne-dEG3-THP: **OH-dEG3-THP** (5.2 g, 10 mmol) was dissolved in dry THF (20 mL). NaH (0.45 g, 60 wt% in mineral oil) was added portionwise with stirring (*Caution: Careful for hydrogen and heat generation*). The reaction mixture was stirred at room temperature for 30 min under N_2 atmosphere. After adding propargyl bromide (1.2 mL, 9.2 M in toluene), the reaction mixture was stirred at room temperature for 3 h. After adding water (100 mL), the product was extracted by ether (100 mL × 2). The organic layer was collected and dried with $MgSO_4$, filtered and concentrated by evaporation. The crude product was purified by column chromatography (SiO_2, Hexane/DCM = 3/2). Yield: 5.2 g (93 %). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 1.45−1.90 (m, 12H), 2.41 (t, J = 2.4 Hz, 1H), 3.45−3.73 (m, 28H), 3.80−3.90 (m, 5H), 4.33 (d, J = 2.4 Hz, 2H), 4.62 (t, J = 3.8 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 19.6, 25.5, 30.7, 57.8, 62.3, 66.8, 70.7, 70.8, 71.0, 71.5, 74.3, 80.4, 99.1.

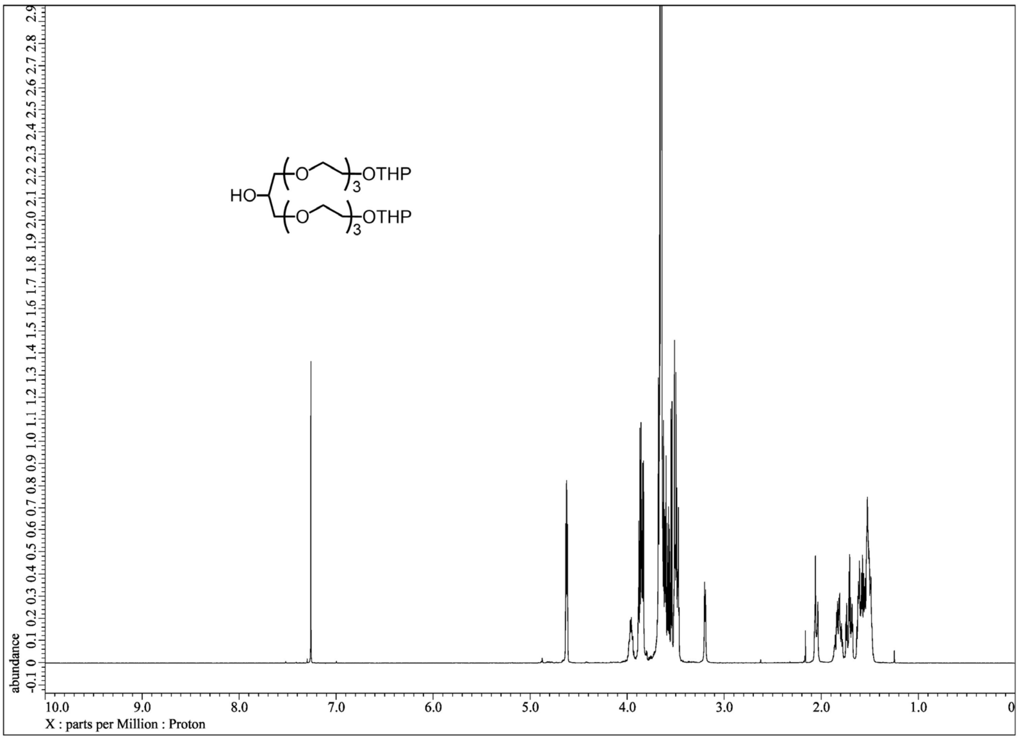
Alkyne-dEG3-OH: **Alkyne-dEG3-THP** (5.0 g, 8.9 mmol) was dissolved in MeOH (200 mL). After adding PPTS (0.25 g, 1.0 mmol), the reaction mixture was heated under reflux at 70 ºC for 16 h. The solvent was removed by evaporation. The residue was directly subjected to column chromatography (SiO_2, DCM/MeOH = 9/1). Yield: 3.0 g (85 %). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 2.41 (t, J = 2.4 Hz, 1H), 2.87 (br, 2H), 3.52−3.75 (m, 28H), 3.87 (m, 1H), 4.32 (d, J = 2.4 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 57.8, 61.8, 70.5, 70.6, 70.8, 70.9, 71.3, 72.7, 74.3, 80.4.

Alkyne-dEG3-Ts: **Alkyne-dEG3-OH** (2.5 g, 6.3 mmol), TEA (2.5 mL, 18 mmol), and DMAP (0.12 g, 1 mmol) were dissolved in dry CH_2Cl_2 (30 mL). After adding TsCl (3.0 g, 16 mmol), the reaction mixture was stirred at room temperature for 16 min under N_2 atmosphere. After adding CH_2Cl_2 (200 mL), the organic solution and 0.1 N HCl aqueous solution (100 mL) were shacked with a separation funnel. The organic layer was collected and dried with $MgSO_4$, filtered and concentrated by evaporation. The crude product was purified by column chromatography (SiO_2, Hexane/AcOEt = 1/4). Yield: 4.2 g (95 %). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 2.40 (t, J = 2.4 Hz, 1H), 2.43 (s, 6H), 3.50−3.64 (m, 20H), 3.67 (t, J = 4.8 Hz, 4H), 3.85 (m, 1H), 4.15 (t, J = 4.8 Hz, 4H), 4.31 (d, J = 2.4 Hz, 2H), 7.33 (d, J = 7.6 Hz, 4H), 7.78 (d, J = 7.7 Hz, 4H); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 21.8, 57.8, 68.8, 69.4, 70.6, 70.9, 71.0, 71.4, 74.3, 76.7, 80.4, 128.1, 129.9, 133.1, 144.9.

Alkyne-dEG3-I: **Alkyne-dEG3-Ts** (4.0 g, 5.7 mmol) and NaI (8.6 g, 57 mmol) were mixed in acetone (40 mL). The reaction mixture was heated under reflux at 60 ºC for 60 h. After evaporation, the residue was dissolved in water (100 mL). The product was extracted by diethyl ether (100 mL × 2). The organic layer was collected and dried with $MgSO_4$, filtered and concentrated by evaporation. The crude product was purified by column chromatography (SiO_2, Hexane/AcOEt = 1/4). Yield: 3.2 g (92 %). ^{1}H NMR (400 MHz, $CDCl_3$): δ = 2.41 (t, J = 2.4 Hz, 1H), 3.26 (t, J = 4.8 Hz, 4H), 3.54−3.68 (m, 20H), 3.75 (t, J = 4.8 Hz, 4H), 3.87 (m, 1H), 4.33 (d, J = 2.4 Hz, 2H); ^{13}C NMR (100 MHz, $CDCl_3$): δ = 3.1, 57.9, 70.4, 70.8, 71.0, 71.5, 72.1, 74.3, 76.8, 80.4.

dEG3-EtIm·NTf$_2$: **Alkyne-dEG3-I** (1.5 g, 2.4 mmol) and EtIm (2.3 mL, 24 mmol) were mixed. After 10 min N_2 bubbling of the solution, the reaction mixture was stirred for 72h at 100 ºC under N_2 atmosphere. The reaction mixture was diluted by distilled water (150 mL). The aqueous solution was washed with CH_2Cl_2 (100 mL × 4) and diethyl ether (100 mL × 2). Li·NTf$_2$ aqueous solution (10.0 g/10mL water) was added to the aqueous solution. The solution was stirred for 1 h at room temperature. The aqueous solution was concentrated to 100 mL by evaporation. The aqueous solution was removed by decantation. The liquid product was dissolved in acetone and dried with $MgSO_4$, filtrated and concentrated by evaporation. Yield: 2.7 g (98 %). ^{1}H NMR (400 MHz, DMSO-d_6): δ = 1.41 (t, J = 7.4 Hz, 6H), 3.38 (t, J = 2.4 Hz, 1H), 3.40−3.57 (m, 20H), 3.68 (m, 1H), 3.77 (t, J = 4.8 Hz, 4H), 4.16−4.24 (overlapping, 6H), 4.33 (t, J = 4.8 Hz, 4H), 7.73 (t, J = 1.6 Hz, 2H), 7.78 (t, J = 1.6 Hz, 2H), 9.11 (s, 2H); ^{13}C NMR (100 MHz, DMSO-d$_6$): δ = 15.1, 44.2, 48.8, 56.6, 68.1, 69.5, 69.6, 70.1, 70.3, 76.1, 76.7, 80.7, 119.5 (q, J = 320.4 Hz), 121.9, 122.8, 136.0.

1.2. ^{1}H and ^{13}C NMR spectra of di-branched derivatives



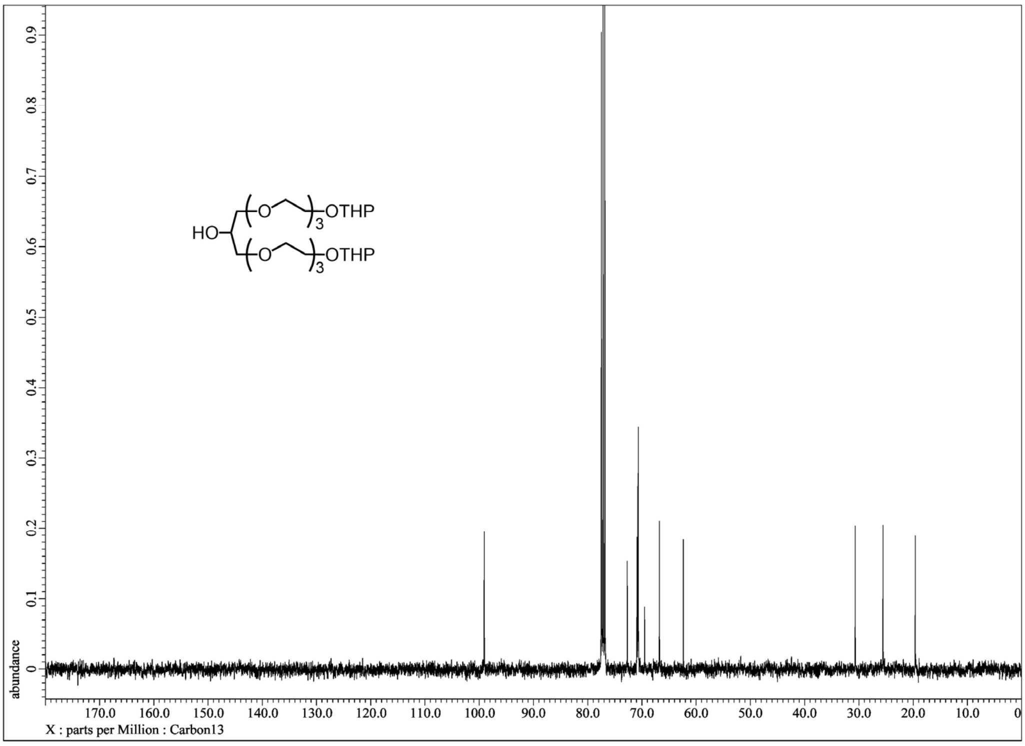


Figure S1 ^{1}H and ^{13}C NMR spectra of **OH-dEG3-THP** ($CDCl_3$)

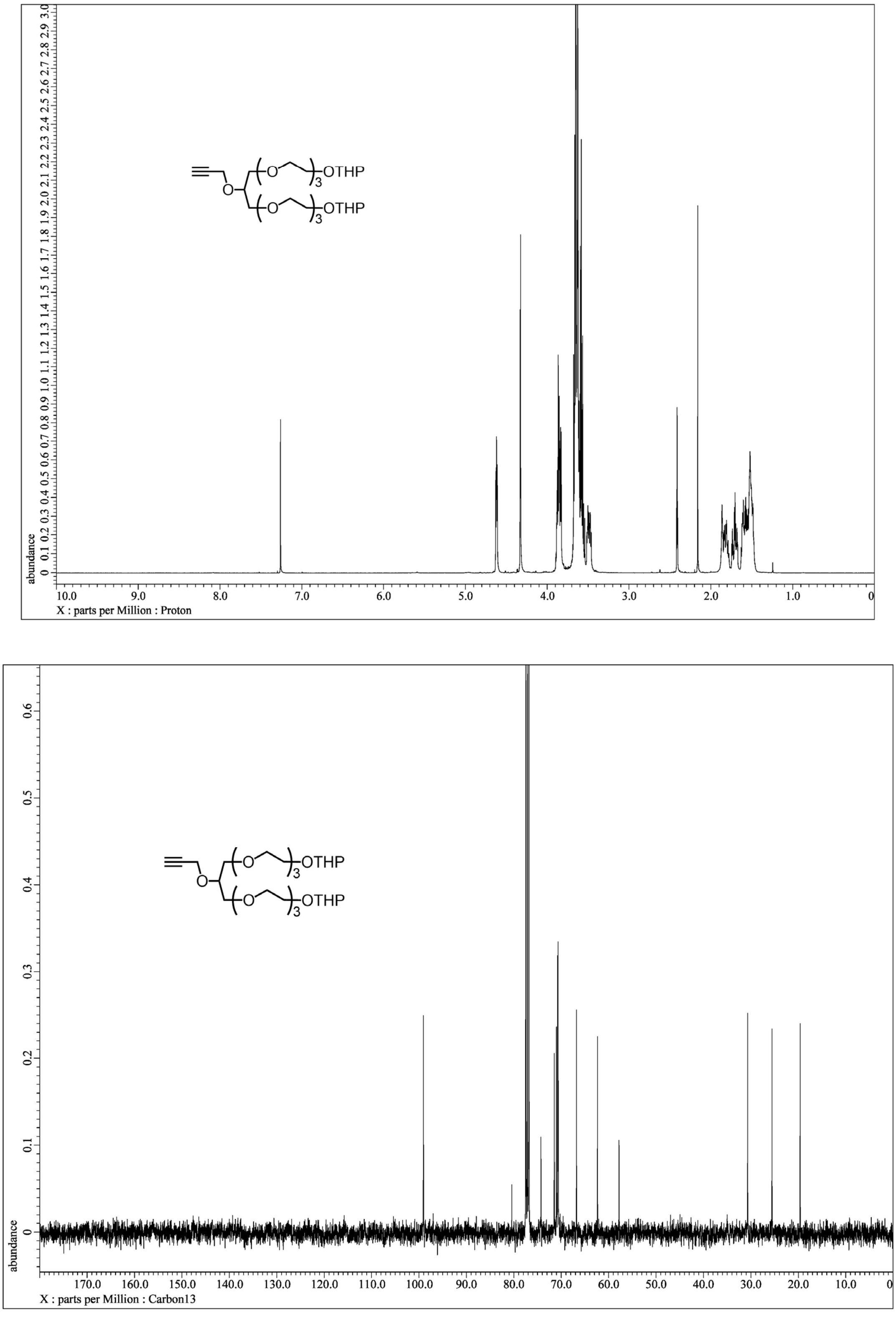
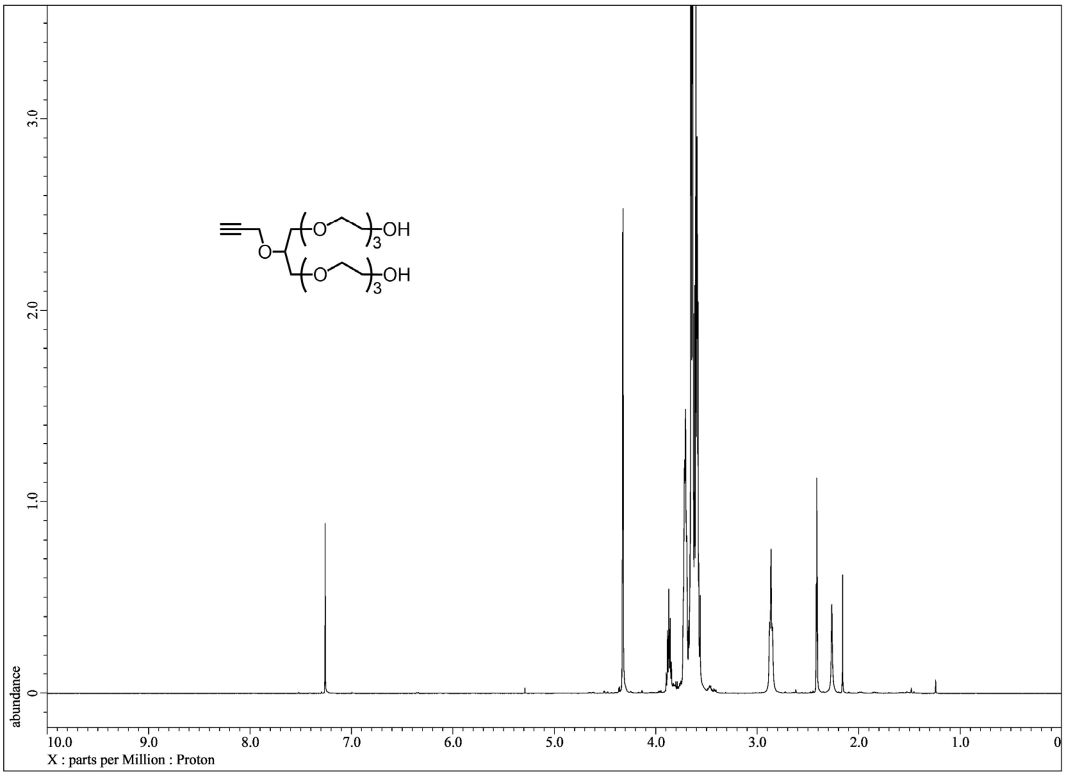


Figure S2 ^{1}H and ^{13}C NMR spectra of **Alkyne-dEG3-THP** ($CDCl_3$)



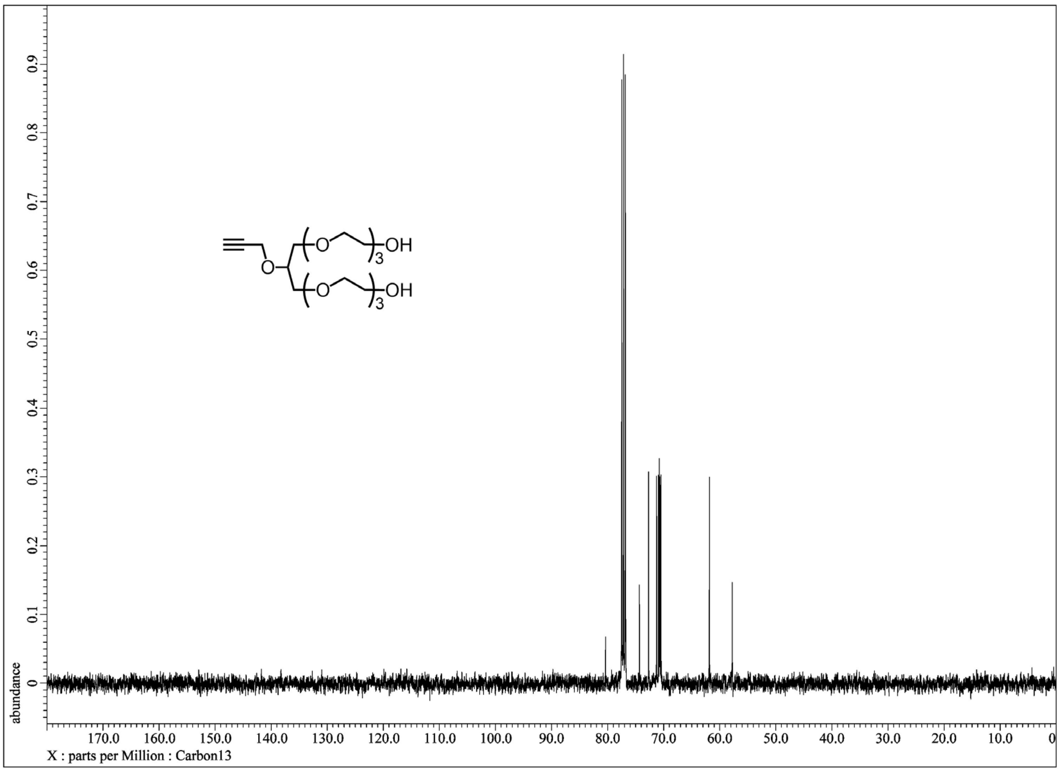
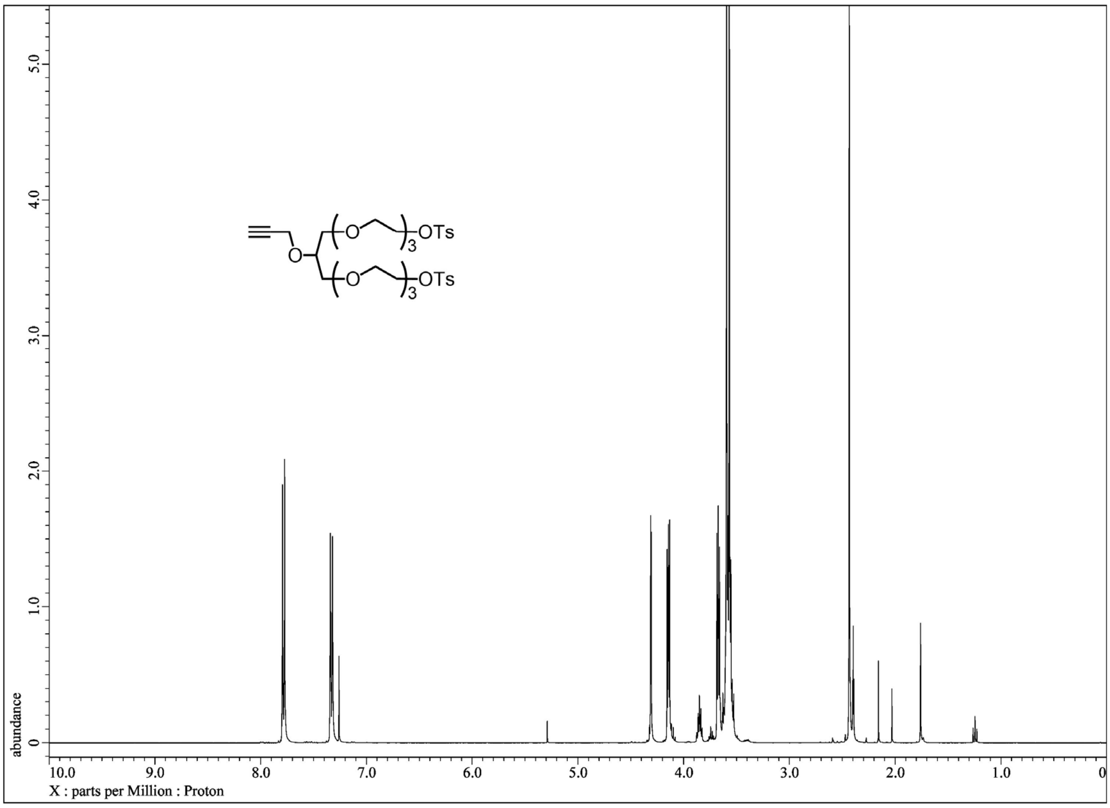


Figure S3 ^{1}H and ^{13}C NMR spectra of **Alkyne-dEG3-OH** ($CDCl_3$)



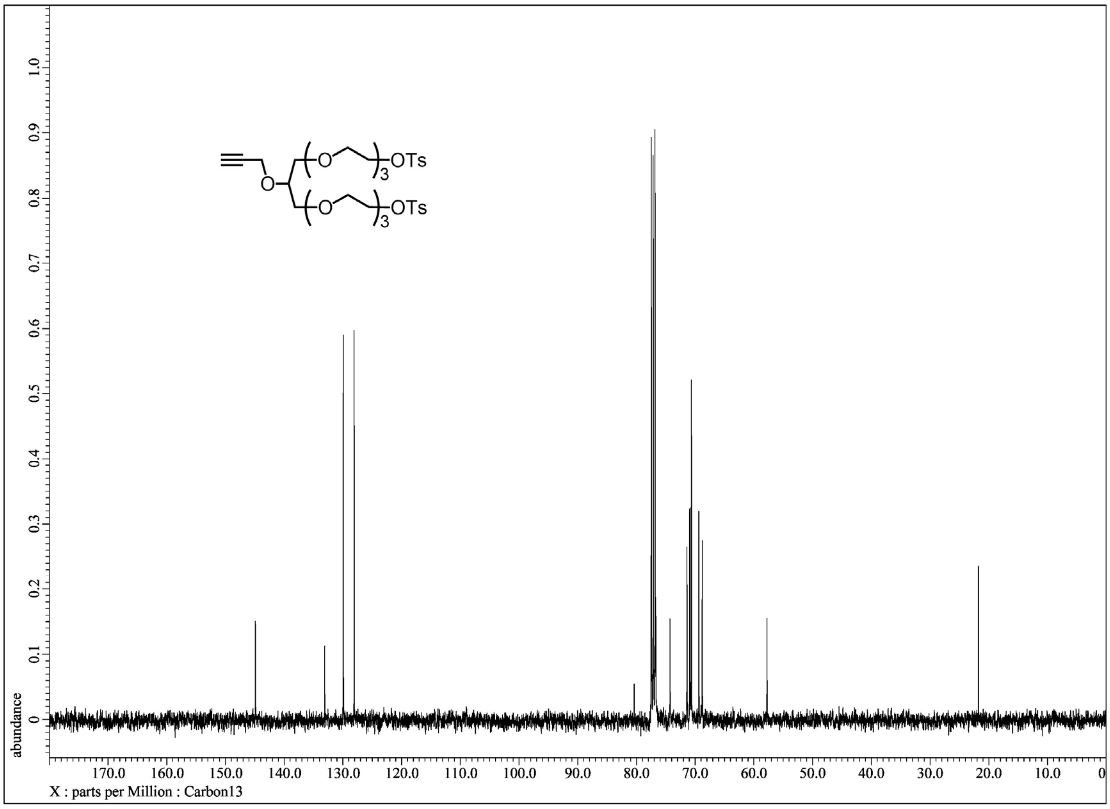


Figure S4 ^{1}H and ^{13}C NMR spectra of **Alkyne-dEG3-Ts** ($CDCl_3$)

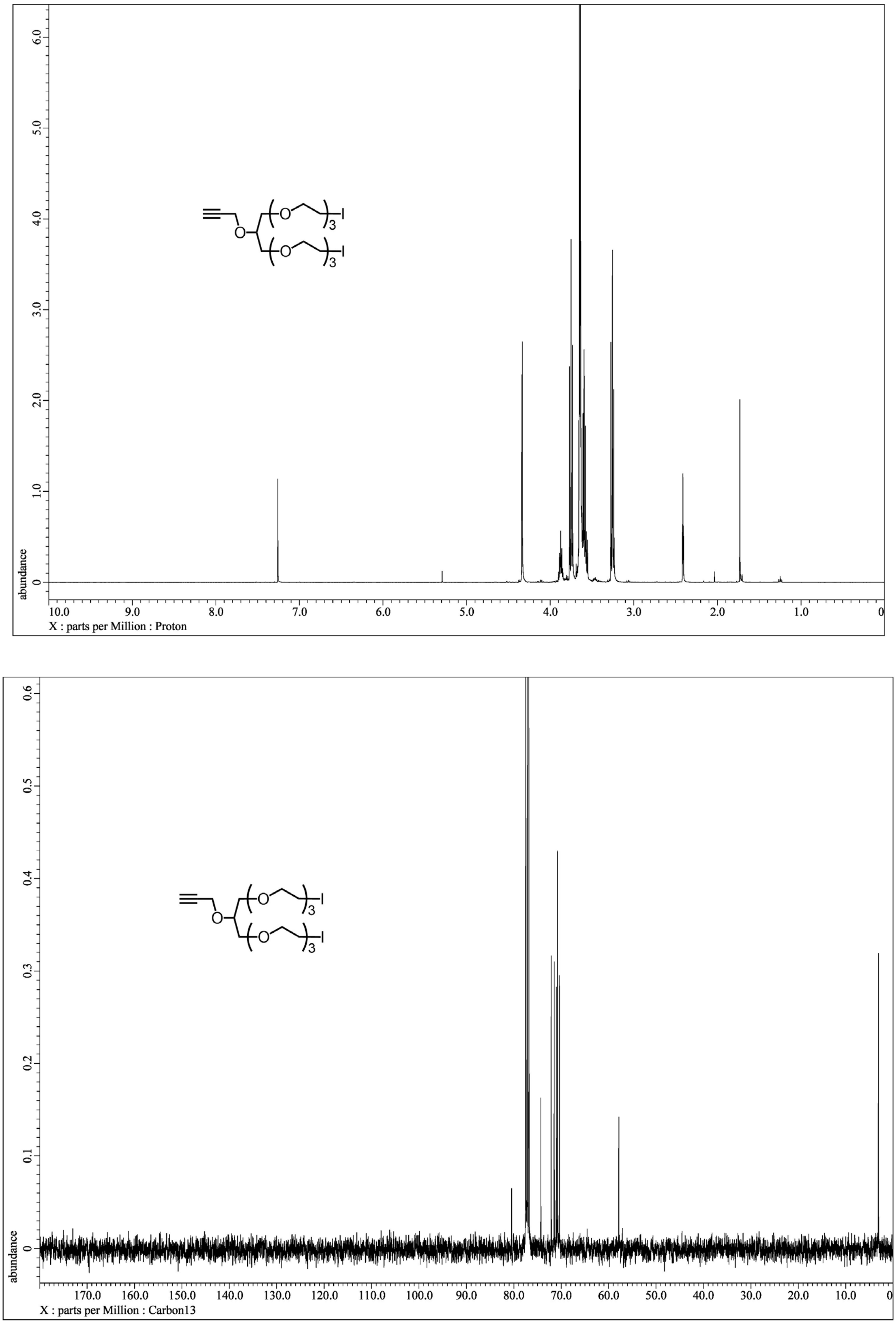


Figure S5 ^{1}H and ^{13}C NMR spectra of **Alkyne-dEG3-I** ($CDCl_3$)

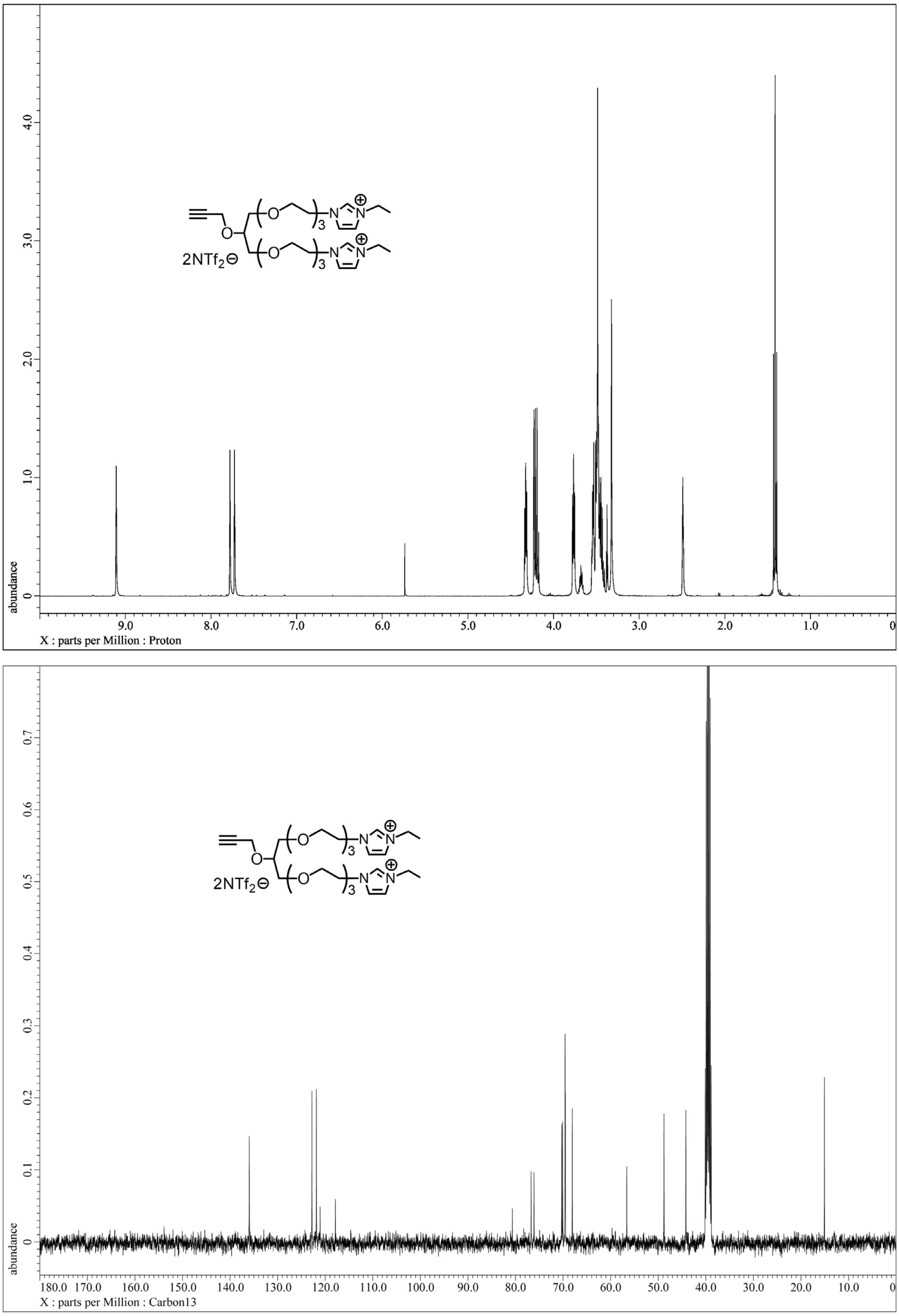
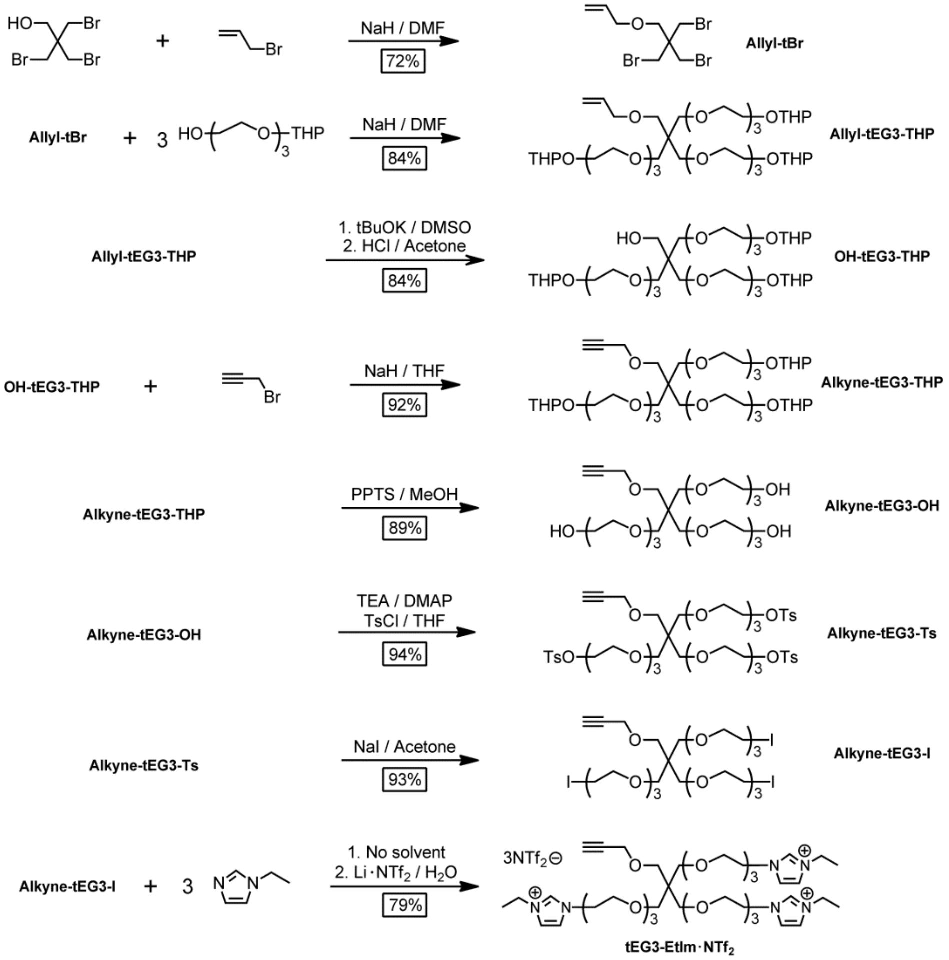


Figure S6 ^{1}H and ^{13}C NMR spectra of **dEG3-EtIm·NTf$_2$** (DMSO-d_6)

2.1. Synthesis of tri-branched cationic alkyne derivative



Scheme S2. Synthesis of tri-branched cationic alkyne derivatives

Allyl-tEG3-THP: THP-protected triethylene glycol (6.0 g, 25.6 mmol) was dissolved in DMF (10 mL). NaH (1.2 g, 60 wt% in mineral oil) was added portionwise with stirring (*Caution: Careful for hydrogen and heat generation*). The reaction mixture was stirred at room temperature for 30 min under N_2 atmosphere. After adding **Allyl-tBr**[S2] (2.0 g, 5.48 mmol), the reaction mixture was stirred at 95 ºC for 24 h. After cooling to room temperature, water (100 mL) was added. The product was extracted with ether (100 mL × 2). The organic layer was collected and dried with $MgSO_4$, filtered and concentrated by evaporation. The crude product was purified by column chromatography (SiO_2, Hexane/Acetone = 3/1). Yield: 3.8 g (84 %). ^{1}H

NMR (400 MHz, CD_3CN): δ = 1.45−1.59 (m, 12H), 1.63−1.83 (m, 6H), 3.37 (s, 2H), 3.40 (s, 6H), 3.42−3.62 (m, 36H), 3.75−3.86 (m, 6H), 3.94 (m, 2H), 4.59 (t, J = 3.6 Hz, 3H), 5.12−5.31 (m, 2H), 5.86−5.96 (m, 1H); ^{13}C NMR (100 MHz, CD_3CN): δ = 20.4, 26.3, 31.5, 46.4, 62.8, 67.5, 69.8, 70.6, 71.0, 71.2, 71.3, 72.0, 72.8, 99.8, 116.4, 136.6.

OH-tEG3-THP: **Allyl-tEG3-THP** (2.5 g, 3.0 mmol) was dissolved in DMSO (10 mL). After adding *t*BuOK (0.5 g, 4.5 mmol), the reaction mixture was stirred at 60 ºC for 30 min. After adding CH_2Cl_2 (100 mL), the solution was washed with 0.1 N HCl aqueous solution (100 mL). The organic layer was collected and dried with $MgSO_4$, filtered and concentrated by evaporation. The recovered residue was dissolved in acetone (25 mL). The solution was cooled to −5 ºC with an ice bath under N_2 atmosphere. After adding acetone (25 mL) containing 0.50 mL conc. HCl aq., the reaction mixture was stirred at −5 ºC for 1 min. The reaction was quenched with triethylamine (0.80 mL). After adding CH_2Cl_2 (100 mL), the solution was washed with 0.1 N $NaHCO_3$ aqueous solution (100 mL). The organic layer was collected and dried with $MgSO_4$, filtered and concentrated by evaporation. The crude product was purified by column chromatography (SiO_2, Hexane/Acetone = 2/3). Yield: 2.0 g (84 %). ^1H NMR (400 MHz, CD_3CN): δ = 1.45−1.59 (m, 12H), 1.63−1.83 (m, 6H), 2.98 (t, J = 6.2 Hz, 1H), 3.42 (s, 6H), 3.43−3.63 (m, 38H), 3.75−3.87 (m, 6H), 4.59 (t, J = 3.4 Hz, 3H); ^{13}C NMR (100 MHz, CD_3CN): δ = 20.4, 26.2, 31.5, 46.2, 62.8, 64.2, 67.5, 70.9, 71.2, 71.2, 71.4, 71.8, 99.8.

Alkyne-tEG3-THP: **OH-tEG3-THP** (2.0 g, 2.5 mmol) was dissolved in dry THF (20 mL). The solution was cooled to −5 ºC with an ice bath under N_2 atmosphere. After adding propargyl bromide (9.2 M in toluene, 0.32 mL) to the solution, NaH (0.15 g, 60 wt% in mineral oil) was added portionwise with stirring (*Caution: Careful for hydrogen and heat generation*). The reaction mixture was stirred for 3 h under N_2 atmosphere. During the reaction, the temperature gradually warmed up to room temperature. After adding water (100 mL), the product was extracted with ether (100 mL × 2). The organic layer was collected and dried with $MgSO_4$, filtered and concentrated by evaporation. The crude product was purified by column chromatography (SiO_2, SNAP50; Hexane/DCM = 7/3). Yield: 1.9 g (92%). ^1H NMR (400 MHz,

CD_3CN): δ = 1.45−1.59 (m, 12H), 1.63−1.83 (m, 6H), 2.73 (t, J = 2.2 Hz, 1H), 3.38 (s, 6H), 3.44 (s, 2H), 3.45−3.62 (m, 36H), 3.75−3.86 (m, 6H), 4.11 (d, J = 2.4 Hz, 2H), 4.59 (t, J = 3.4 Hz, 3H); ^{13}C NMR (100 MHz, CD_3CN): δ = 21.4, 27.2, 32.4, 47.1, 60.2, 63.7, 68.4, 70.8, 71.4, 71.9, 72.1, 72.2, 72.9, 76.5, 82.1, 100.7.

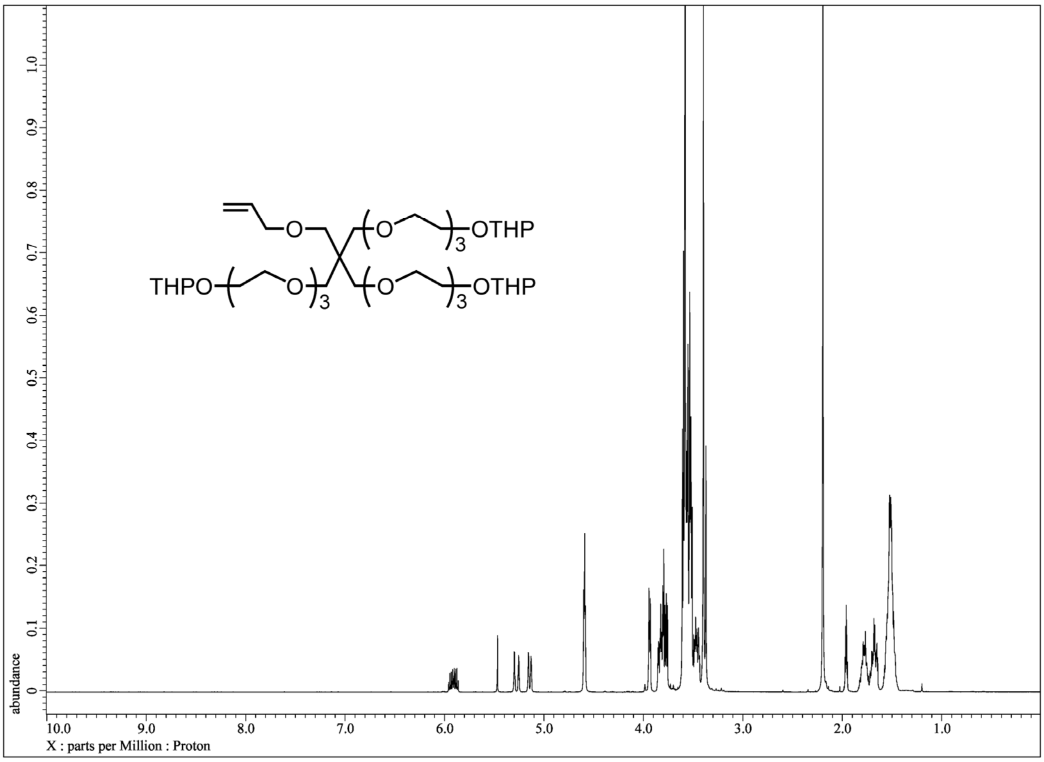
Alkyne-tEG3-OH: **Alkyne-tEG3-THP** (1.8 g, 2.2 mmol) and PPTS (0.30 g, 1.2 mmol) were dissolved in MeOH (180 mL). The reaction mixture was stirred at 80 ºC for 16 h under N_2 atmosphere. After removing the solvent by evaporation, the residue was directly subjected to column chromatography (SiO_2, SNAP50; DCM/MeOH = 9/1). Yield: 1.1 g (89%). ^1H NMR (400 MHz, CD_3CN): δ = 2.72 (t, J = 2.4 Hz, 1H), 2.90 (br, 3H), 3.40 (s, 6H), 3.45 (s, 2H), 3.49−3.64 (m, 36H), 4.11 (d, J = 2.4 Hz, 2H); ^{13}C NMR (100 MHz, CD_3CN): δ = 46.1, 59.3, 62.0, 69.9, 70.5, 71.0, 71.1, 71.3, 71.9, 73.3, 75.6, 81.2.

Alkyne-tEG3-Ts: **Alkyne-tEG3-OH** (1.0 g, 1.8 mmol), TEA (1.0 mL, 7.2 mmol) and DMAP (50 mg, 0.41 mmol) were dissolved in dry CH_2Cl_2 (10 mL). After adding TsCl (1.3 g, 6.8 mmol), the reaction mixture was stirred at room temperature for 16 h under N_2 atmosphere. After diluting the reaction mixture with CH_2Cl_2 (100 mL), the solution was washed with 0.1 N HCl aqueous solution (100 mL) and 0.1 N $NaHCO_3$ aqueous solution (100 mL). The organic layer was dried with $MgSO_4$, filtered and concentrated by evaporation. The crude product was purified by column chromatography (SiO_2, DCM/Acetone = 4/1). Yield: 1.7 g (94%). ^1H NMR (400 MHz, CD_3CN): δ = 2.45 (s, 9H), 2.70 (t, J = 2.4 Hz, 1H), 3.36 (s, 6H), 3.42 (s, 2H), 3.45−3.55 (m, 24H), 3.62 (t, J = 4.6 Hz, 6H), 4.09 (d, J = 2.4 Hz, 2H), 4.12 (t, J = 4.6 Hz, 6H), 7.45 (d, J = 8.0 Hz, 6H), 7.80 (d, J = 8.0 Hz, 6H); ^{13}C NMR (100 MHz, CD_3CN): δ = 21.7, 30.9, 46.1, 59.3, 69.2, 69.8, 70.5, 71.0, 71.1, 71.2, 71.9, 75.6, 81.2, 128.8, 131.0, 133.8, 146.4.

Alkyne-tEG3-I: **Alkyne-tEG3-Ts** (1.6 g, 1.5 mmol) and NaI (3.5 g, 23 mmol) were dissolved in dry acetone (35 mL). The reaction mixture was stirred at 60 ºC for 60 h under N_2 atmosphere. After removing the solvent by evaporation, the residue was dissolved in CH_2Cl_2 (100 mL), and washed with water (100 mL). The organic layer was dried with $MgSO_4$, filtered and concentrated by evaporation. The crude product was purified by column chromatography (SiO_2, DCM/Acetone = 4/1). Yield: 1.3 g (93%). ^1H NMR (400 MHz, CD_3CN): δ= 2.72 (t, J = 2.4 Hz, 1H), 3.32 (t, J = 6.8 Hz, 6H), 3.39 (s, 6H), 3.45 (s, 2H), 3.51−3.64 (m, 24H), 3.73 (t, J = 6.8 Hz, 6H), 4.12 (d, J = 2.4 Hz, 2H); ^{13}C NMR (100 MHz, CD_3CN): δ= 5.0, 46.2, 59.3, 69.9, 70.5, 70.8, 71.0, 71.2, 71.9, 72.4, 75.6, 81.2.

tEG3-EtIm·NTf_2: **Alkyne-tEG3-I** (0.90 g, 1.0 mmol) and 1-ethyl-imidazole (1.5 mL, 15.6 mmol) were stirred at 100 ºC for 72 h under N_2 atmosphere. The reaction mixture was diluted by distilled water (150 mL). The aqueous solution was washed with CH_2Cl_2 (100 mL × 4) and diethyl ether (100 mL × 2). Li·NTf_2 aqueous solution (5.0 g/10 mL water) was added to the aqueous solution. The solution was stirred for 1 h at room temperature. The aqueous solution was concentrated to 100 mL by evaporation. The aqueous solution was removed by decantation. The liquid product was dissolved in acetone and dried with $MgSO_4$, filtrated and concentrated by evaporation. Yield: 1.3 g (79 %). ^1H NMR (400 MHz, CD_3CN): δ= 1.49 (t, J = 7.4 Hz, 9H), 2.71 (t, J = 2.2 Hz, 1H), 3.37 (s, 6H), 3.42 (s, 2H), 3.49−3.63 (m, 24H), 3.80 (t, J = 4.8 Hz, 6H), 4.09 (d, J = 2.2 Hz, 2H), 4.20 (q, J = 7.4 Hz, 6H), 4.29 (t, J = 4.8 Hz, 6H), 7.42 (t, J = 1.6 Hz, 3H), 7.46 (t, J = 1.6 Hz, 3H), 8.55 (s, 3H); 15.5, 45.9, 46.2, 50.5, 59.3, 69.2, 69.9, 70.6, 70.9, 71.0, 71.1, 71.9, 75.6, 81.2, 120.9 (q, J = 319 Hz), 122.7, 124.1, 136.5.

2.2. ^{1}H and ^{13}C NMR spectra of tri-branched derivatives



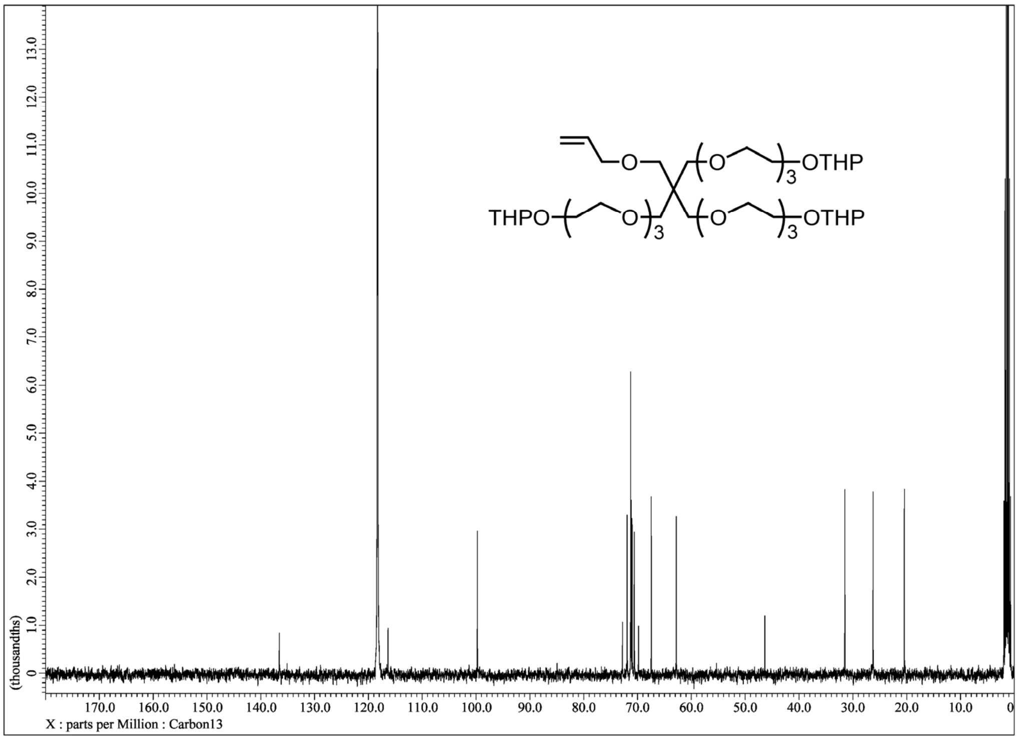
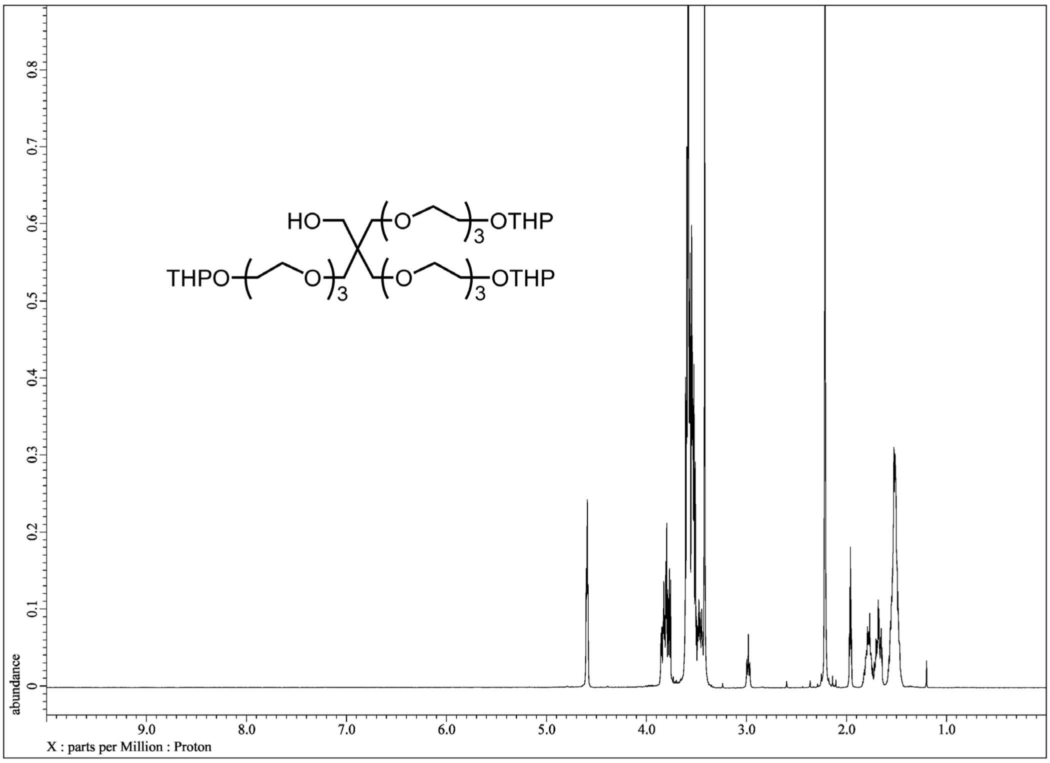


Figure S7 ^{1}H and ^{13}C NMR spectra of **Allyl-tEG3-THP** (CD_3CN)



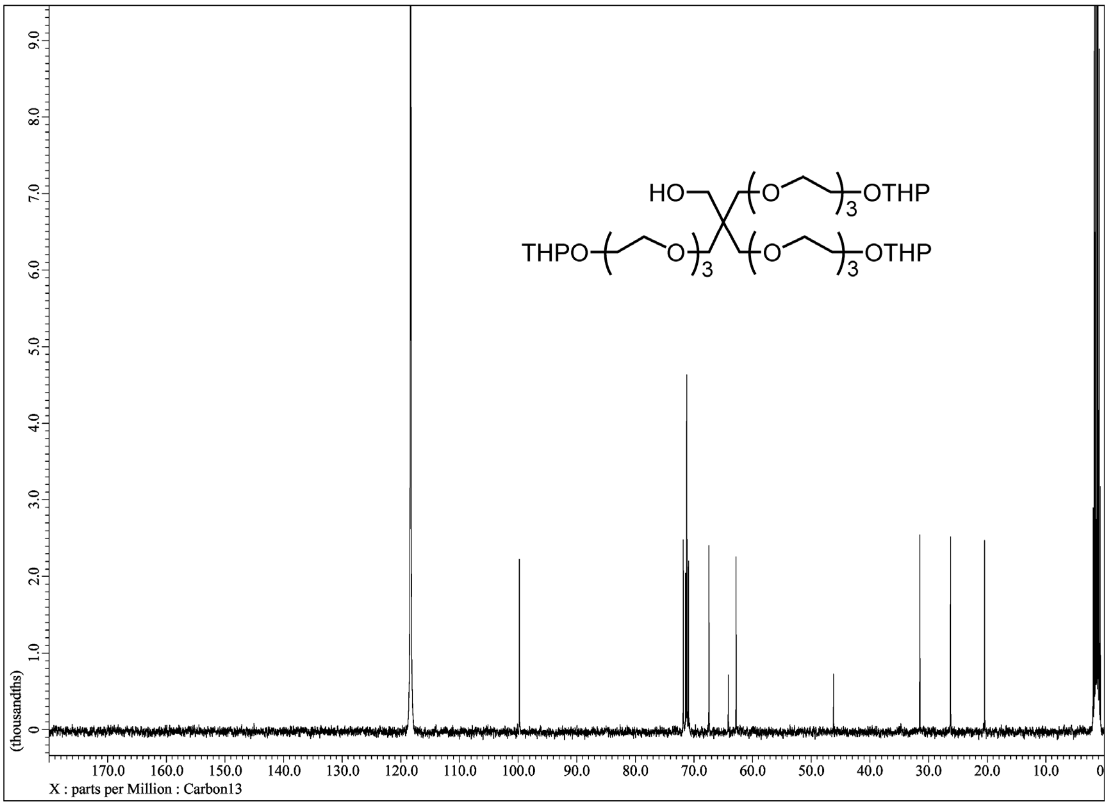


Figure S8 ^{1}H and ^{13}C NMR spectra of **OH-tEG3-THP** (CD$_3$CN)

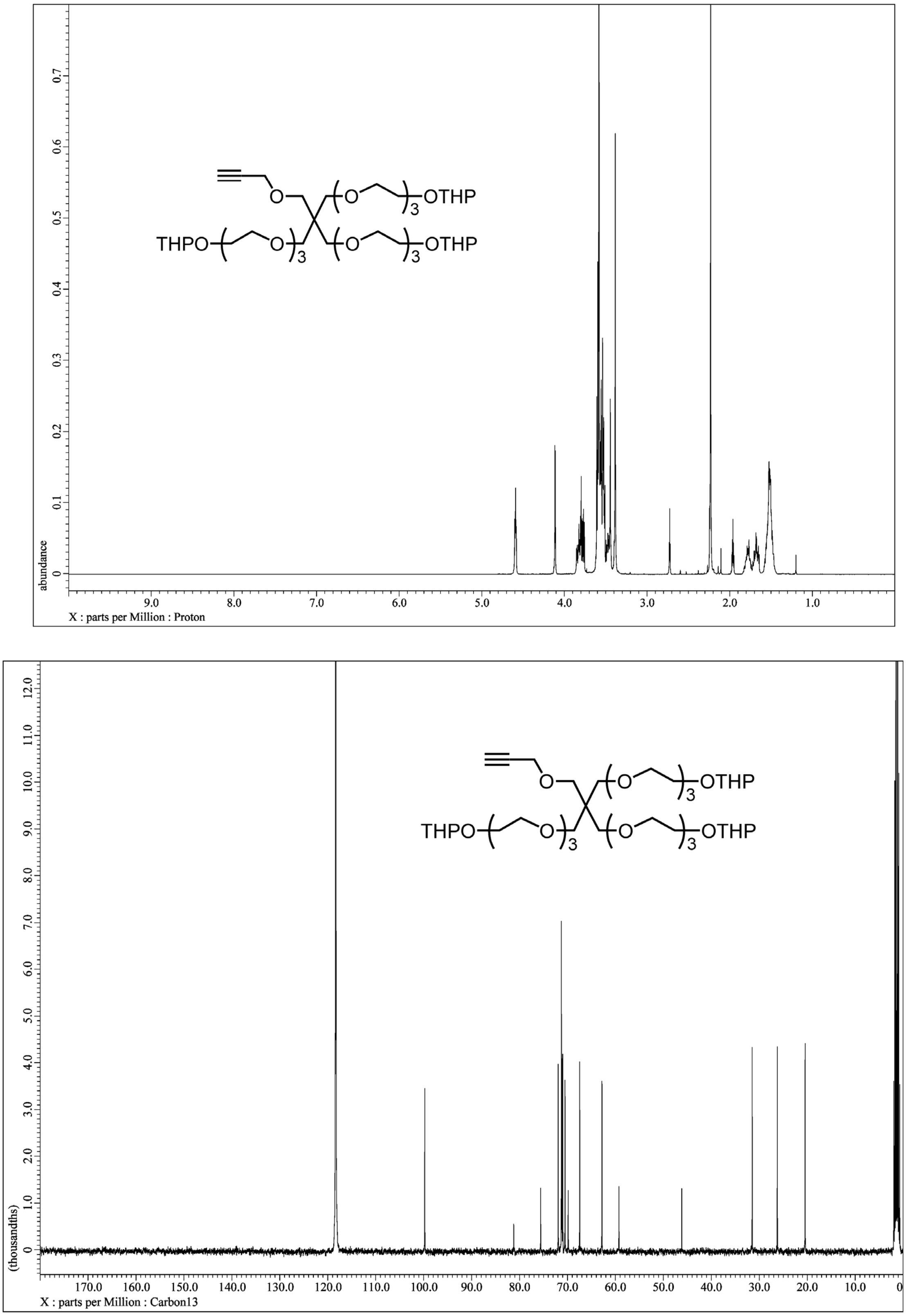


Figure S9 ^{1}H and ^{13}C NMR spectra of **Alkyne-tEG3-THP** (CD_3CN)

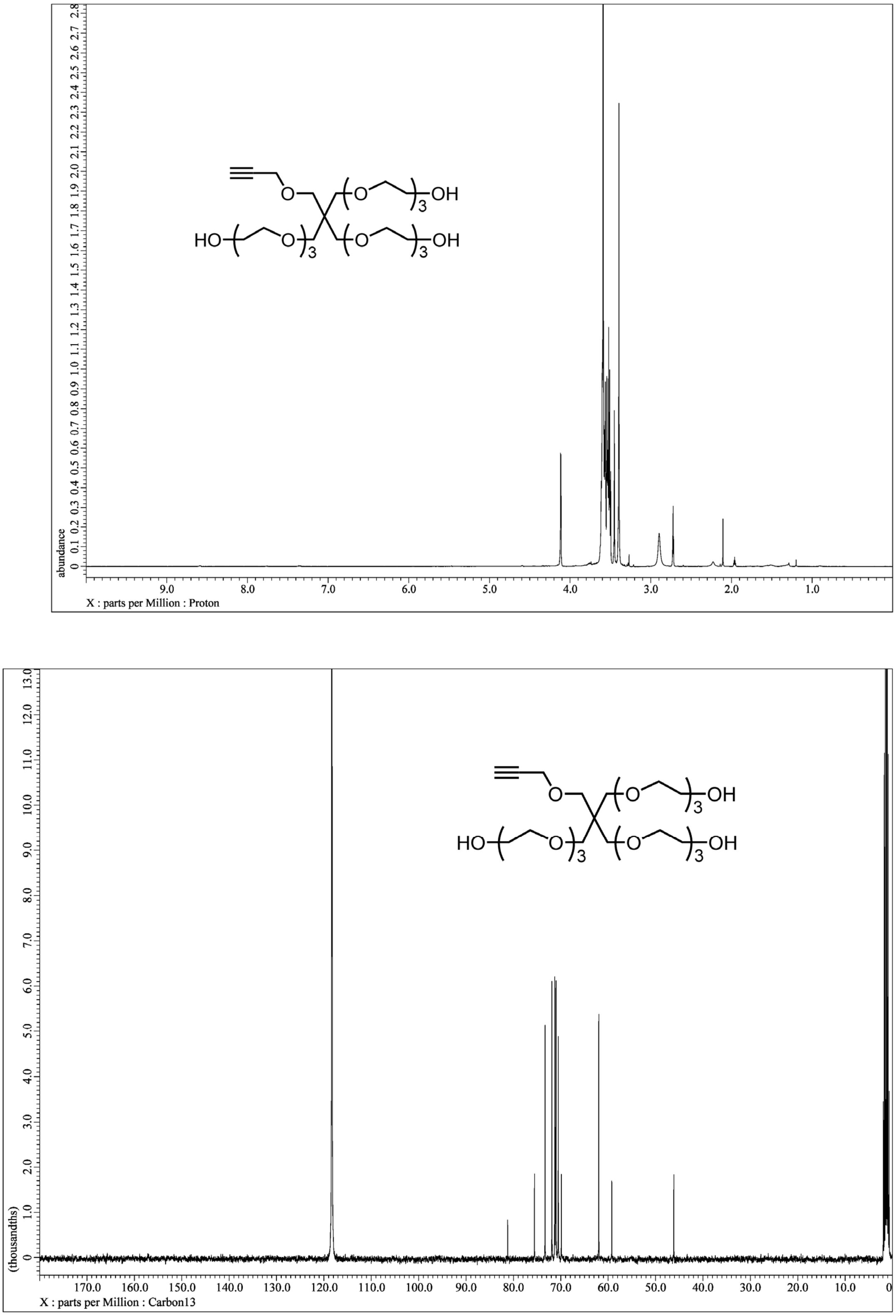
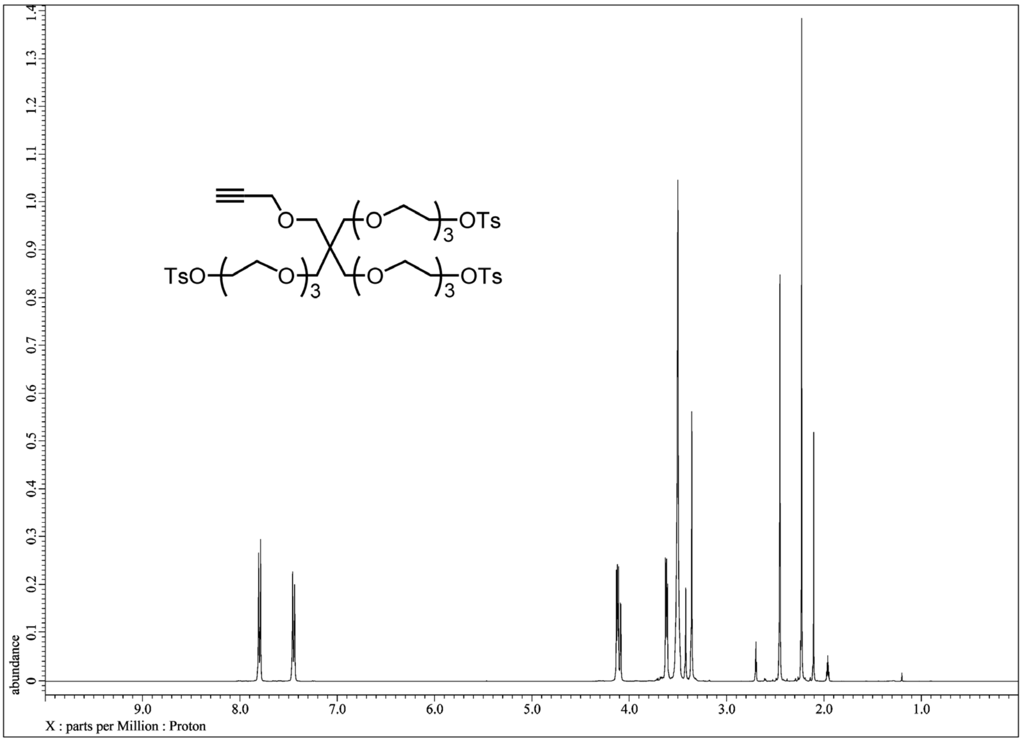


Figure S10 ^{1}H and ^{13}C NMR spectra of **Alkyne-tEG3-OH** (CD_3CN)



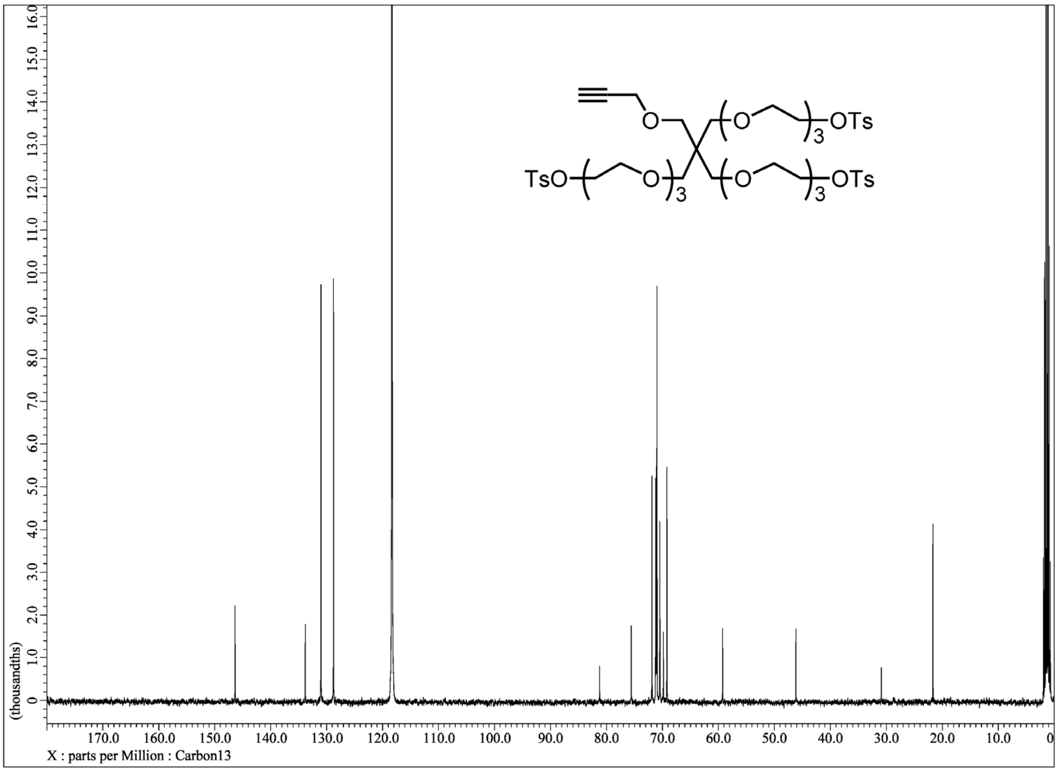
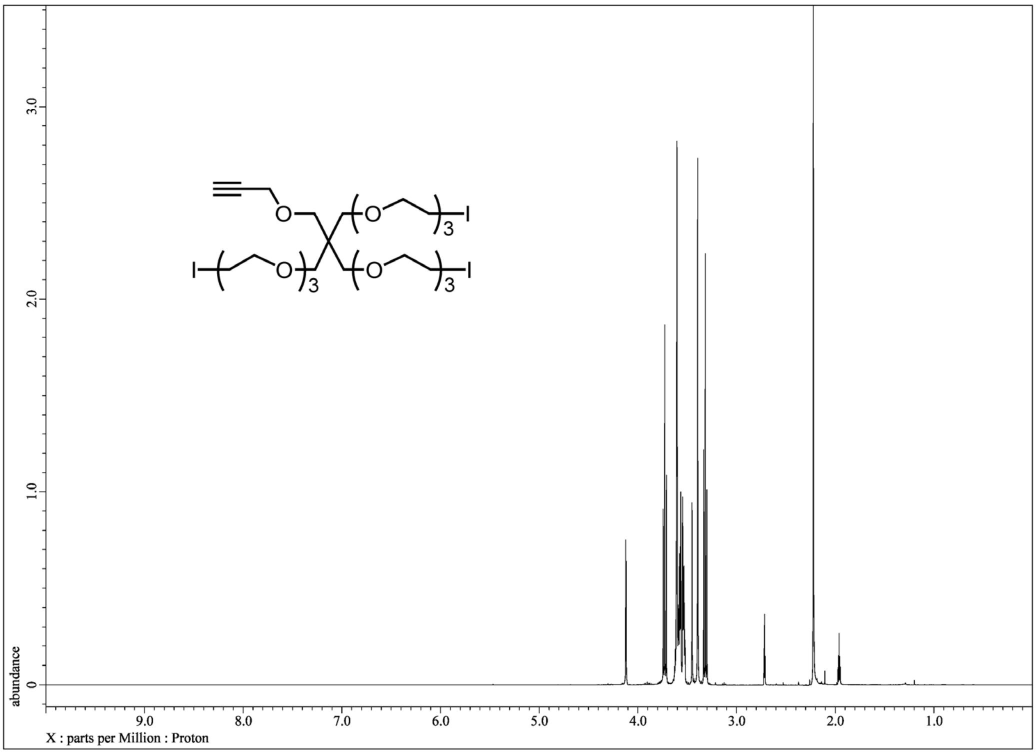


Figure S11 ^{1}H and ^{13}C NMR spectra of **Alkyne-tEG3-Ts** (CD_3CN)



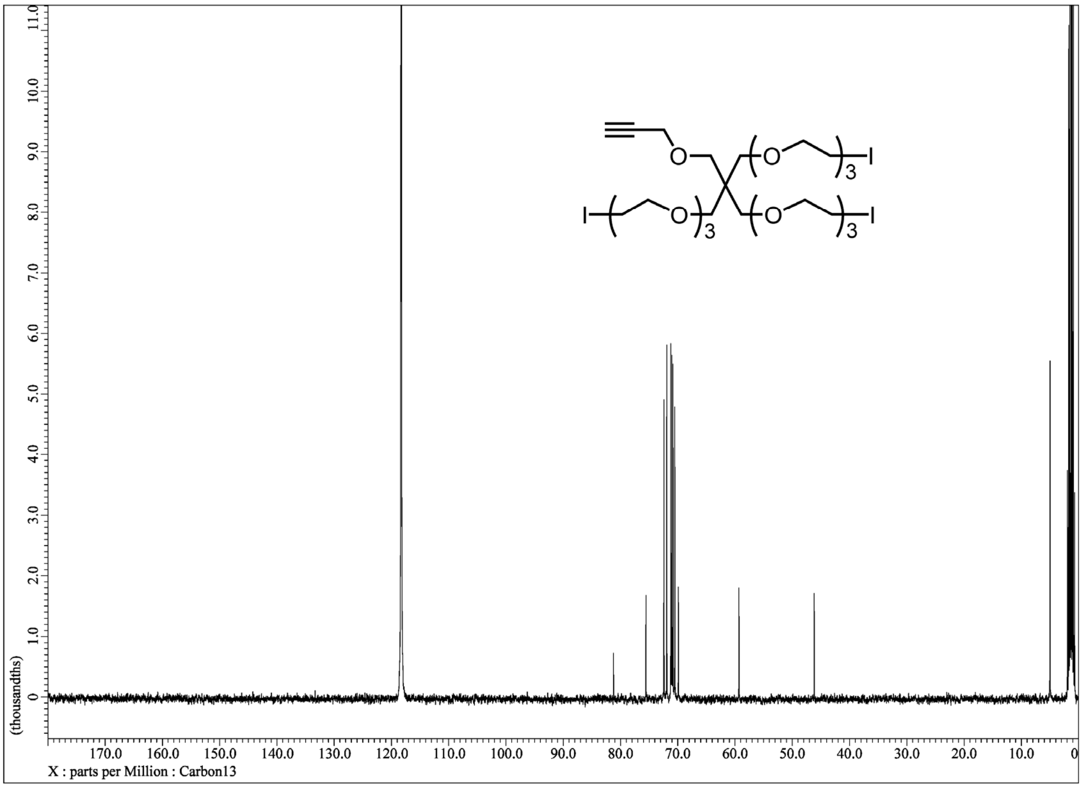


Figure S12 ^{1}H and ^{13}C NMR spectra of **Alkyne-tEG3-I** (CD_3CN)

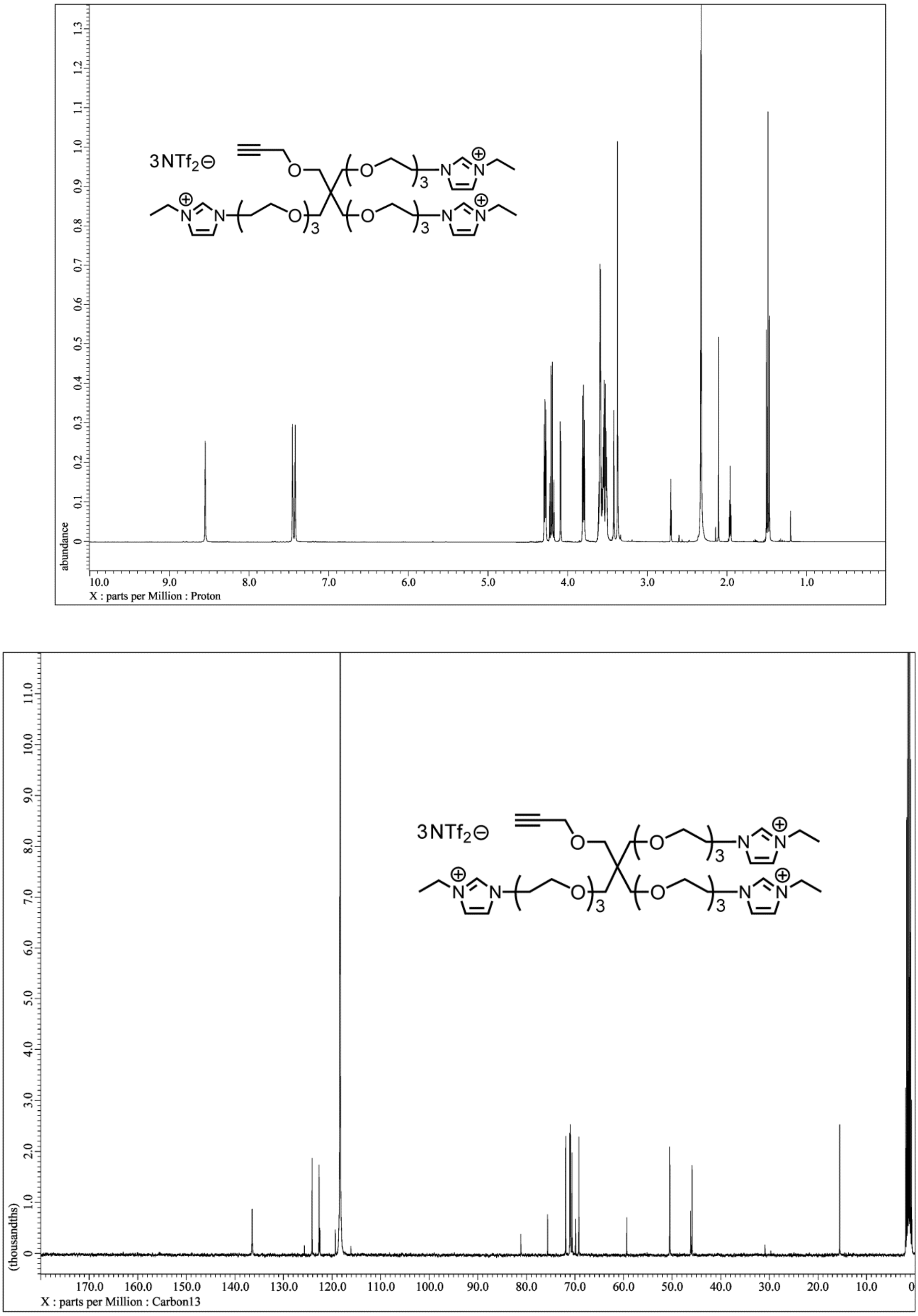


Figure S13 ^{1}H and ^{13}C NMR spectra of **tEG3-EtIm·NTf$_2$** (CD_3CN)

3. IR spectra of GAP and cationic GTPs

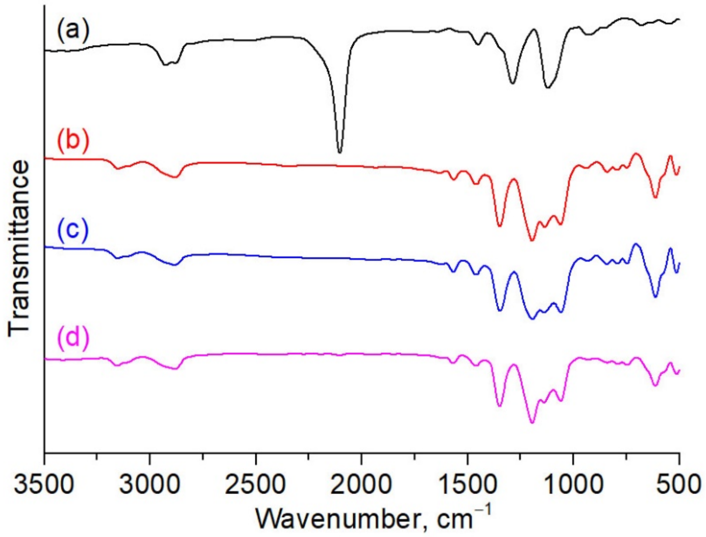


Figure S14 FT-IR spectra of GAP and cationic GTPs. (a) GAP, (b) GTP-mEG4-EtIm·NTf$_2$, (c) GTP-dEG3-EtIm· NTf$_2$, and (d) GTP-tEG3-EtIm·NTf$_2$ (KBr pellet)

4. ^{1}H NMR spetra of cationic GTPs

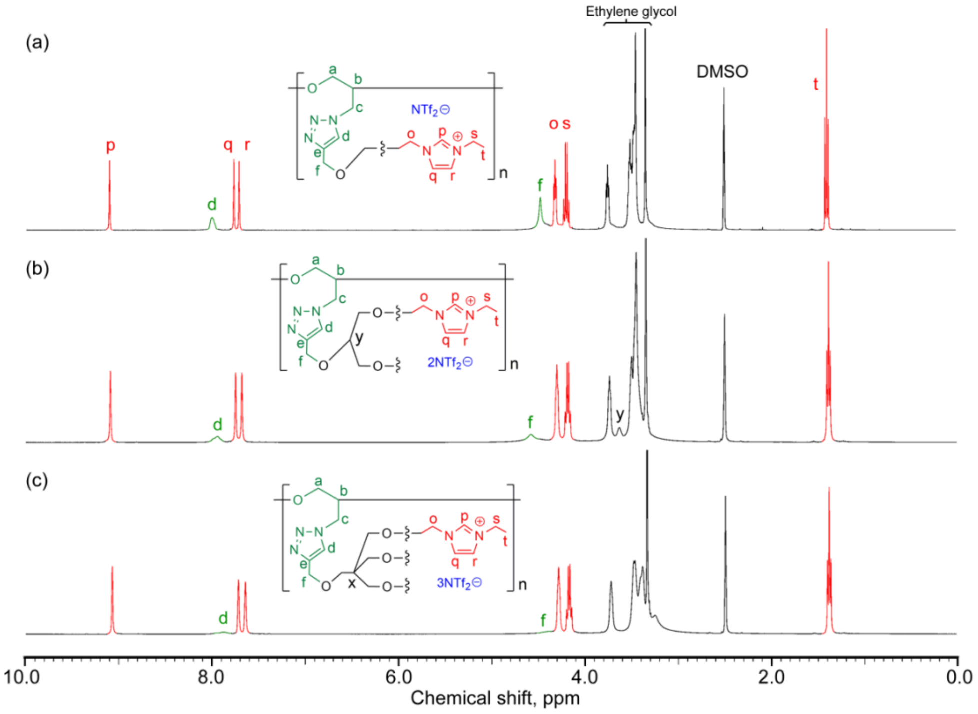


Figure S15 ^{1}H NMR spectra of cationic GTPs (a) GTP-mEG4-EtIm·NTf$_2$, (b) GTP-dEG3-EtIm· NTf$_2$, and (c) GTP-tEG3-EtIm·NTf$_2$ (DMSO-d_6)

5. SEC data

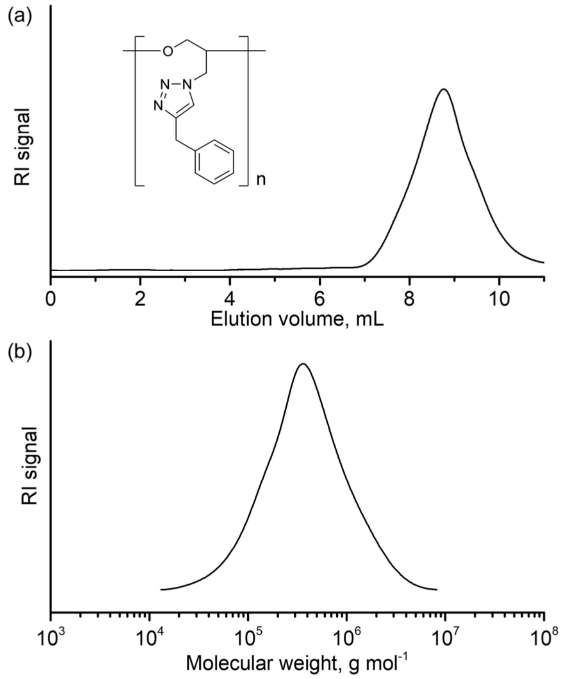


Figure S16. (a) SEC trace of GTP-Bz (50 °C, 0.01 M Li·NTf$_2$ in DMF). Chemical structure of GTP-Bz is depicted. (b) Molecular weight distribution curve of GTP-Bz.

6. DSC data

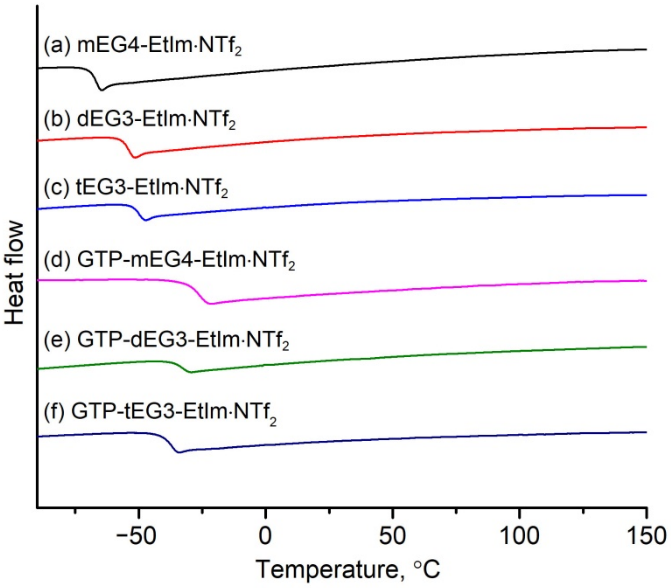


Figure S17. DSC traces of cationic alkynes and GTPs. Heating rate 10 °C min^{-1}.

7. TGA data

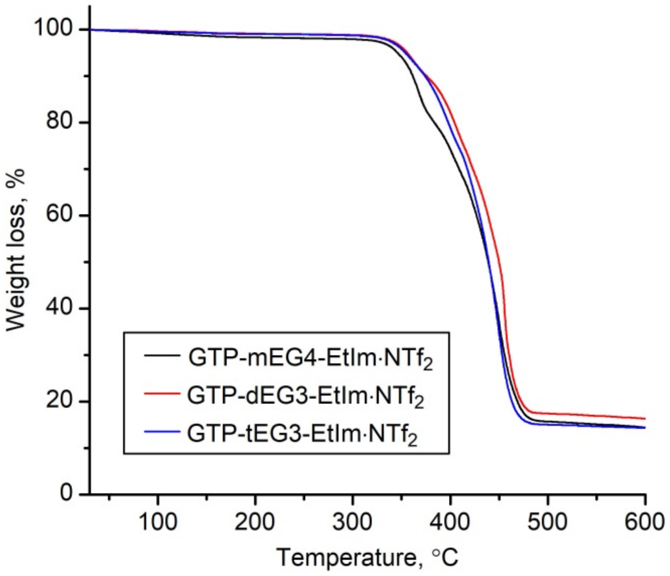


Figure S18. TGA traces of cationic GTPs. N_2 atmosphere. Heating rate 10 °C min^{-1}.

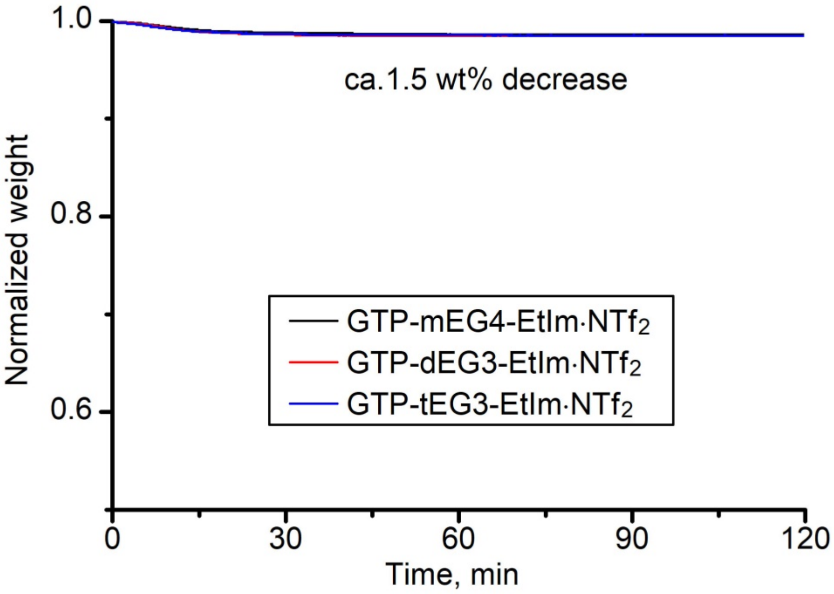


Figure S19. TGA traces of cationic GTPs at 120°C. N_2 atmosphere.

8. Impedance data

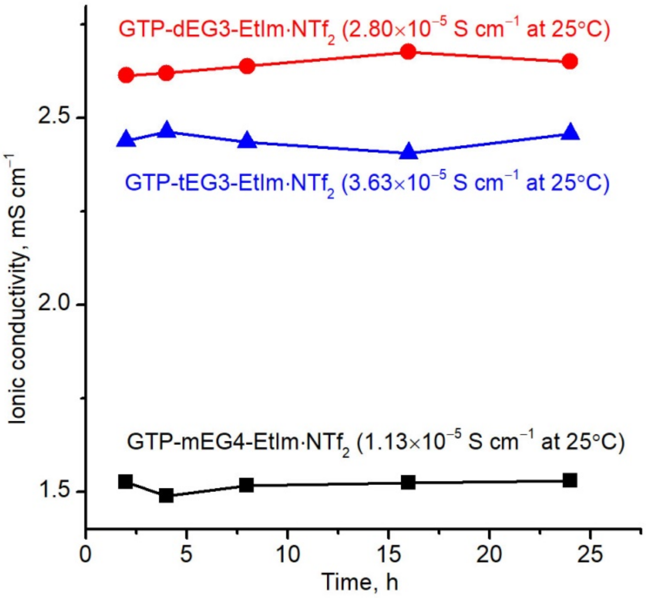


Figure S20. Conductivity change under 120 °C for 24h.
Values in parenthesis are conductivity at 25 °C after dry process under 120 °C for 24h.

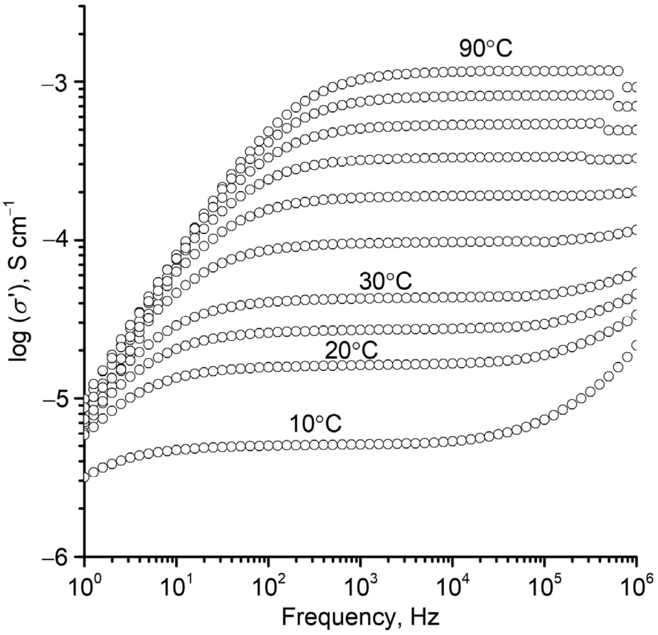


Figure S21. Conductivity vs frequency plot at temperatures range from 90 to 10 °C for **GTP-dEG3-EtIm·NTf$_2$**. Anhydrous condition.

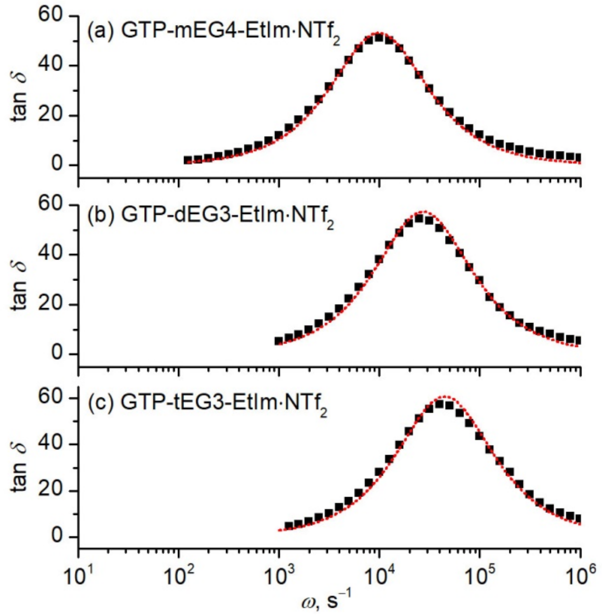


Figure S22. The plot of tan d vs angular frequency at 25 °C under dry condition. The dot curves were obtained from fitting by equation (3).

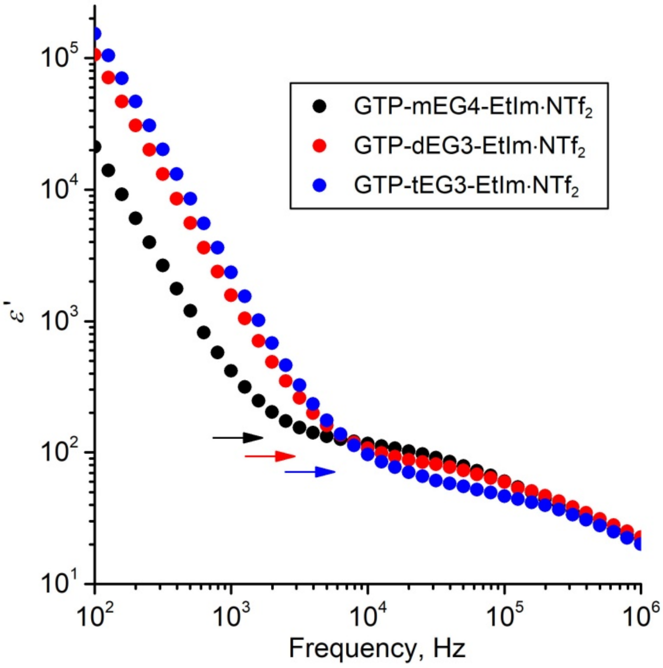


Figure S23. Dielectric constant vs frequency plot at 25°C. Arrows indicate calculated static dielectric constant by equation (6).

Table S1. Ionic conductivity of GTP derivatives (Average ± standard error, *n* = 4)

T °C	GTP-mEG4-EtIm·NTf_2 S cm^{-1}	GTP-dEG3-EtIm·NTf_2 S cm^{-1}	GTP-tEG3-EtIm·NTf_2 S cm^{-1}
90	$7.39 \pm 0.31 \times 10^{-4}$	$1.12 \pm 0.04 \times 10^{-3}$	$1.13 \pm 0.01 \times 10^{-3}$
80	$5.02 \pm 0.22 \times 10^{-4}$	$7.81 \pm 0.24 \times 10^{-4}$	$8.09 \pm 0.06 \times 10^{-4}$
70	$3.09 \pm 0.12 \times 10^{-4}$	$5.19 \pm 0.15 \times 10^{-4}$	$5.53 \pm 0.04 \times 10^{-4}$
60	$1.86 \pm 0.08 \times 10^{-4}$	$3.20 \pm 0.07 \times 10^{-4}$	$3.55 \pm 0.06 \times 10^{-4}$
50	$1.01 \pm 0.05 \times 10^{-4}$	$1.85 \pm 0.04 \times 10^{-4}$	$2.12 \pm 0.03 \times 10^{-4}$
40	$4.77 \pm 0.23 \times 10^{-5}$	$9.64 \pm 0.23 \times 10^{-5}$	$1.15 \pm 0.02 \times 10^{-4}$
30	$1.93 \pm 0.09 \times 10^{-5}$	$4.34 \pm 0.08 \times 10^{-5}$	$5.53 \pm 0.11 \times 10^{-5}$
20	$6.34 \pm 0.31 \times 10^{-6}$	$1.64 \pm 0.03 \times 10^{-5}$	$2.25 \pm 0.05 \times 10^{-5}$
10	$1.68 \pm 0.06 \times 10^{-6}$	$5.39 \pm 0.11 \times 10^{-6}$	$8.16 \pm 0.35 \times 10^{-6}$

Table S2. Conducting ion concentration of GTP derivatives (Average ± standard error, *n* = 4)

T °C	GTP-mEG4-EtIm·NTf_2 cm^{-3}	GTP-dEG3-EtIm·NTf_2 cm^{-3}	GTP-tEG3-EtIm·NTf_2 cm^{-3}
30	$1.12 \pm 0.07 \times 10^{18}$	$1.21 \pm 0.05 \times 10^{18}$	$1.32 \pm 0.08 \times 10^{18}$
20	$9.30 \pm 0.61 \times 10^{17}$	$9.82 \pm 0.25 \times 10^{17}$	$1.04 \pm 0.06 \times 10^{18}$
10	$7.55 \pm 0.57 \times 10^{17}$	$8.05 \pm 0.30 \times 10^{17}$	$8.99 \pm 0.65 \times 10^{17}$

Table S3. Conducting ion mobility of GTP derivatives (Average ± standard error, *n* = 4)

T °C	GTP-mEG4-EtIm·NTf_2 cm^2 V^{-1} s^{-1}	GTP-dEG3-EtIm·NTf_2 cm^2 V^{-1} s^{-1}	GTP-tEG3-EtIm·NTf_2 cm^2 V^{-1} s^{-1}
30	$1.09 \pm 0.11 \times 10^{-4}$	$2.22 \pm 0.05 \times 10^{-4}$	$2.68 \pm 0.10 \times 10^{-4}$
20	$4.32 \pm 0.41 \times 10^{-5}$	$1.04 \pm 0.01 \times 10^{-4}$	$1.39 \pm 0.05 \times 10^{-4}$
10	$1.42 \pm 0.14 \times 10^{-5}$	$4.14 \pm 0.13 \times 10^{-5}$	$5.87 \pm 0.12 \times 10^{-5}$

Table S4. Static dielectric constant of GTP derivatives (Average ± standard error, *n* = 4)

T °C	GTP-mEG4-EtIm·NTf$_2$	GTP-dEG3-EtIm·NTf$_2$	GTP-tEG3-EtIm·NTf$_2$
30	129 ± 5	87.0 ± 1.6	62.8 ± 1.5
20	138 ± 5	98.1 ± 2.4	77.9 ± 2.4
10	148 ± 8	108 ± 2	86.2 ± 3.8

9. References

S1. F. A. Loiseau, K. K. Hii and A. M. Hill, *J. Org. Chem.*, 2004, **69**, 639.

S2. D. Lee, H. Y. Jung and M. J. Park, *ACS Macro Lett.*, 2018, **7**, 1046.